Combustion Research Facility News

Structure, propagation of triple flames with detailed chemistry studied

Tarek Echekki and Jacqueline Chen have been studying the structure, propagation, and stabilization mechanisms of a methanol-air triple flame in a scalar mixing layer using direct numerical simulation with detailed chemistry and transport models.

Triple flames arise in a number of practical configurations where the fuel and oxidizer are partially pre-mixed. From the fuel-rich to the fuel-lean sides of the mixture, rich and lean premixed branches form and span the range of mixture flammability. Behind them, a diffusion-flame branch is stabilized at the triple point (the locus where the three branches meet) and is fed by the `excess' fuel and oxidizer. The triple flame structure provides a mechanism for diffusion flame stabilization that may be present in lifted jet diffusion flames.

An account of detailed chemistry and transport effects (16 species, 51 reversible reactions) reveals some of the interesting features of triple flames that are overlooked by simple reaction rate and transport models.

In the present methanol-air computation, Tarek and Jackie observed that the primary fuel, methanol, is consumed entirely in the premixed branches of the flame (see Figure 1), and is converted to more stable fuels, H_2 and CO, that are consumed in the trailing diffusion flame. The excess oxidizer, oxygen, on the other hand does survive past the lean premixed branch (Figure 1).

Another interesting feature of triple flames that is central to the stabilization of the diffusion branch is the complex nature of the triple point. It is characterized by strong diffusion fluxes and enhanced radical production amplified by the curvature of the leading edge of the flame.

Figure 1 shows the reaction rate of H_2 in the flame. The enhanced H_2 production is due primarily to its focusing towards its oxidation zone by virtue of the leading edge's curvature. This results in enhanced radical production through key chain-branching steps involving H_2 in the triple region. This enhancement serves as an anchoring mechanism for the diffusion branch. It also results in an enhancement of the triple flame's propagation speed relative to the unburnt gas. However, Tarek and Jackie find that the enhancement in the flame's propagation speed is still primarily governed by hydrodynamic effects that are adequately described by simpler chemical and transport models with heat release.

Tarek and Jackie propose a new parameterization of the triple flame structure in terms of two phase space parameters that span the entire range of reactedness and premixedness: mixture fraction and temperature. This parameterization highlights the different topologies of the flame that, in a turbulent flow, may be obscured in physical coordinates. Figure 2 shows the reaction rate of H_2, previously shown in physical space, in the phase plane. Peak O_2 consumption rate is used to demarcate the three branches of the flame.

In practical situations, triple flames are present in turbulent flow fields or are subject to shear, such as in the case of turbulent jet diffusion flames. In the future, the team plans to include mean shear effects on triple flames. The study will provide further insight into the complex structure of triple flames in more realistic conditions.
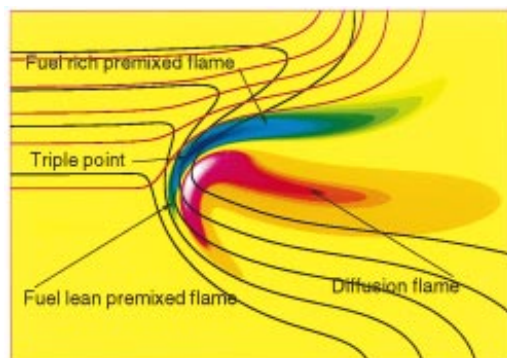

Figure 1. Hydrogen molecule reaction rate. Blue and white correspond to maxima in production and consumption rates, respectively (same for Figure 2). Methanol and oxygen mass fraction isocontours are denoted by red and black contour lines, respectively.
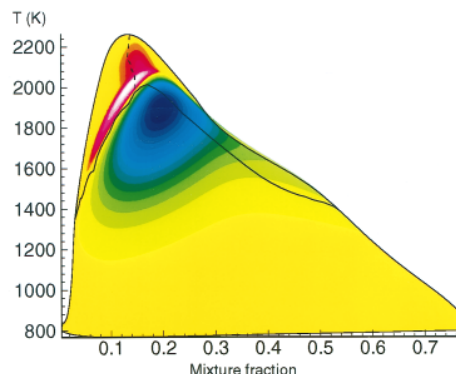

Figure 2. Hydrogen molecule reaction rate in temperature and mixture fraction phase space. Maximum oxygen consumption rate in the premixed and diffusion branches is denoted by black solid and dashed lines, respectively. Stoichiometric mixture fraction is 0.136.

Tunable diode lasers measure transient emissions from a rotary kiln

A team composed of Sarah Allendorf, Howard Johnsen, Dave Ottesen, Chris Shaddix, David Rosenberg (Cornell University co-op student), and Gary Hubbard (Hubbard Associates), recently performed field trials at a pilot-scale rotary kiln incinerator operated by the U.S. Environmental Protection Agency (EPA) at Research Triangle Park, North Carolina. This project, funded through the Environmental Technologies Initiative and managed by the EPA, was designed to evaluate the potential for using near-infrared tunable diode lasers (TDLs) as fast-response combustion performance indicators.

In many cases, both the emission performance and the economics of operation of hazardous waste incinerators are limited by the occurrence of fuel-rich system upsets and the resultant formation of "puffs" of unburned pollutant species. These transient emissions may be minimized by injecting air or pure oxygen into a secondary combustion chamber when an appropriate sensor in the primary chamber indicates the presence of a puff and determines its magnitude. However, the application of such feed-forward emission control logic is at present limited by the lack of a commercially available, fast-response puff sensor.

For the rotary kiln trials, two wavelength-modulated TDLs that span carbon monoxide (CO) and methane (CH_4) vibrational absorption lines at approximately 1.6 μm were used. Two in-situ, line-of-sight measurement locations were accessed, one just downstream of the rotary kiln section and one near the end of the secondary combustion chamber. In addition, a long-pass optical cell was used to perform more sensitive TDL measurements on extractively sampled gases.

Figure 1 compares in-situ TDL measurements of CO and methane with those from conventional extractive-sampling continuous emission monitors (CEMs) during "steady," non-sooty operation of the kiln. The methane TDL yields sensitive, quantitative results, whereas the CO TDL signal suffers from weak CO line strengths at the 1320 K kiln temperature and interference from carbon dioxide and water hot-band lines.

Figure 2 shows typical TDL measurements during a series of puffs generated in the rotary kiln by the time-controlled injection of a mixture of toluene and methylene chloride. Extensive attenuation of the laser beam inside the kiln (due to soot formation) prevented the in-situ measurement of CO and methane, but this loss in transmissivity in itself provided a sensitive, rapid puff indicator. TDL analysis of extractively sampled, filtered gases provided a quantitative measure of CO and methane concentrations.

Previous research at the EPA has demonstrated that measurements of CO, methane, and soot levels during puffs can be used to determine the amount of air or oxygen addition required for complete burn-out of the puff. The results from the TDL experiments suggest that a combination of in-situ and extractive gas cell TDL measurements holds the most promise for a fast-responding, quantitative puff-sensor system that can accommodate upset conditions with a variety of soot loadings. ✎
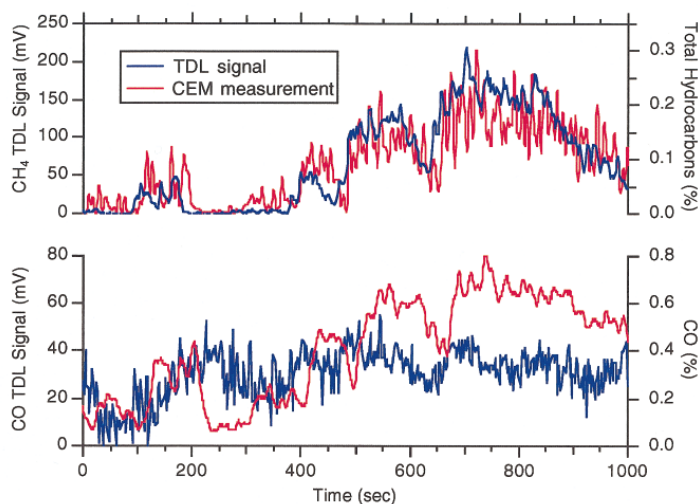

Figure 1. Time record of TDL signals and corresponding CEM measurements during operation of the kiln pilot flame of natural gas/air under slightly fuel-rich conditions. Note the faster response time of the TDL measurements.
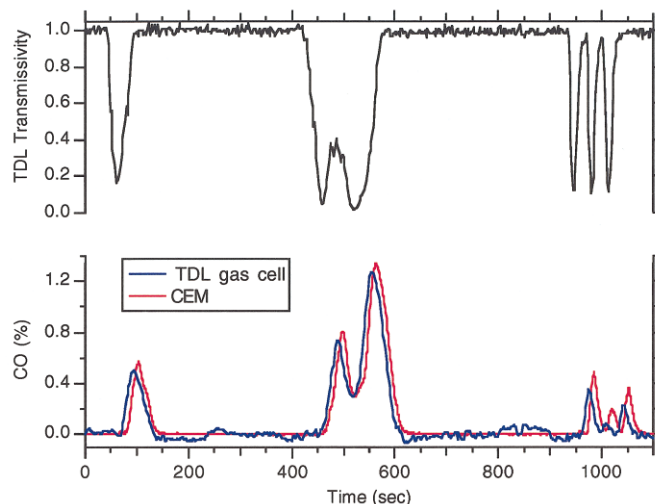

Figure 2. Time record of TDL signals and CO CEM measurements from the kiln sampling location during a series of puffs. The TDL gas cell measurement has a long response time in this experiment because of its coupling into the CEM gas sampling system.

Lina Prada is a graduate student at the University of Zaragoza, Spain. She spent last fall at the CRF working with Jim Miller on developing a comprehensive chemical kinetic model for NO_x reburning.

Hans Heinrich Carstensen (seated), now at Exxon Research, spent last year working with Joe Durant on aspects of nitrogen chemistry.

Edward Coy was a postdoctoral appointee working with John Dec on laser induced incandescence of soot and with Jay Keller on lean premixed combustion of hydrogen. He has joined Hughes STX at Edwards Air Force Base, California.

Cornell Co-op Program participants visit

Each year Sandia cooperates in an integrated education plan that puts Cornell University juniors at Sandia for a semester, back at Cornell for a semester, and returns them to Sandia the following summer for research in their chosen field. The Cornell program has existed since 1991 and has accommodated 45 students. This fall, seven students have been visiting Sandia, four in the CRF.

(Shown clockwise from top right) David Rosenberg, has been working with Sarah Allendorf, Dave Ottesen, Gary Hubbard, and Howard Johnsen to develop laser-based sensors for gas species, concentration, and temperature measurements for basic oxygen steelmaking process control.

Jessica Wickham has assisted Steve Rice and visiting scientist Eric Croiset in the supercritical oxidation project on hydrogen peroxide thermal dissociation rates as well as supporting Dick Steeper and Jason Aiken on water-gas shift kinetics at hydrothermal conditions.

Candace Morey has been a member of Larry Baxter's Multifuel Combustor Laboratory team, working with Steve Buckley on the reapplication of energetic materials as boiler fuels and Helle Junker (Elsamprojekt, Denmark) and Allen Robinson on studies of ash deposition during biomass and coal combustion.

Dana Adams worked with Chris Shaddix and Melissa Lunden on the biomass fuel oils and coal kinetics projects.

CRF on the WorldWide Web

Please visit the CRF home page at http://www.ca.sandia.gov/CRF/. Read What's HOT at the CRF for updates on the CRFII building project (featured in February), information on the Advisory Board (January), and exciting projects such as our joint effort in waste cleanup with Belarus (December).

Infrared absorption provides sensitive probe for chemical kinetics

The study of chemical kinetics plays a critical role in understanding and modeling complex systems such as combustion and can provide fundamental information about important chemical species. Craig Taatjes and Jeff Pilgrim have been applying sensitive infrared absorption techniques to temperature-dependent kinetics studies.

Infrared absorption is a general technique for probing chemical species, but is less sensitive than many other optical probing methods. A simple yet powerful way to increase the sensitivity of absorption measurements is to use multiple passes of the probe laser.

The multipass apparatus developed here is based on the Herriott design, which uses off-axis paths in a spherical resonator (Figure 1). The kinetics experiments use a pulsed pump laser to initiate the reaction being studied and follow the reaction by time-resolved absorption of a continuous infrared probe. Combining the Herriott cell with an on-axis pump laser allows the pump-probe overlap to be confined to the center of the reactor, where the temperature is precisely controlled.

This apparatus has recently been used to study the reaction of Cl atoms with ethylene, C_2H_4. This reaction can proceed by addition to form C_2H_4Cl or by hydrogen abstraction to form HCl and a vinyl radical (C_2H_3). The abstraction is related to the heat of formation of the vinyl radical, an important species in hydrocarbon combustion. A discrepancy has existed between kinetics measurements, which yield $\Delta H°_{f,298}(C_2H_3)$ of ~67 kcal/mol, and spectroscopic studies, which imply $\Delta H°_{f,298}(C_2H_3)$ of ~72 kcal/mol.

Extracting temperature-dependent rate coefficients for the abstraction channel in Cl + C_2H_4 is experimentally challenging because of the competing addition. However, the abstraction can be clearly separated by measuring at higher temperatures, where the addition is less important, and by carefully determining the HCl yield.

The rate coefficients for the hydrogen abstraction are shown in Figure 2. Using published measurements of the reverse reaction, HCl + C_2H_3, these kinetic data establish an experimental heat of formation for the vinyl radical of 71.6 ±0.3 kcal/mol, in agreement with spectroscopic determinations.

Craig and Jeff continue to use the multipass apparatus to investigate temperature-dependent kinetics of important combustion species, including methyl (CH_3) and formyl (HCO) radicals. This work is supported by the Division of Chemical Sciences, the Office of Basic Energy Sciences, the U.S. Department of Energy.
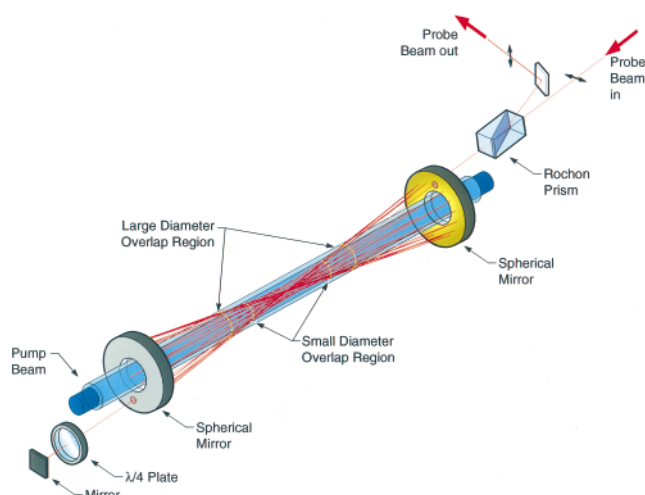

Figure 1. The multiple-pass resonator. The extent of the pump-probe overlap can be adjusted by changing the diameter of the pump laser.
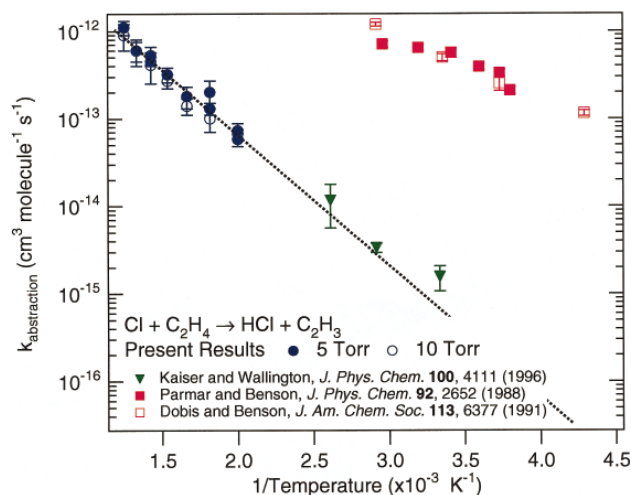

Figure 2. An Arrhenius plot of the abstraction rate data. The Arrhenius expression obtained is given as the dashed line in the figure.